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(54) Title: EXHAUST LINE FOR AN INTERNAL COMBUSTION ENGINE

(57) Abstract: An exhaust system for an internal combustion engine comprises a catalysed particulate filter comprising a NOx absorbent capable of absorbing NOx contained in an exhaust gas when the composition of the exhaust gas is lambda > 1, and capable of releasing the NOx absorbed in the NOx component when the exhaust gas composition is 1 ≥ lambda, characterised in that the exhaust system further comprises a catalyst capable of oxidising NO to NO_2 at least when the air-fuel ration of the exhaust gas is lean.

EXHAUST LINE FOR AN INTERNAL COMBUSTION ENGINE

The present invention relates to an exhaust system for a lean burn internal combustion engine, and in one illustrative embodiment, to an exhaust system for a diesel engine.

Lean burn internal combustion engines, such as diesel engines and gasoline engines, produce a number of pollutants including carbon monoxide (CO), unburnt hydrocarbon (HC), particulate matter (PM) and nitrogen oxides (NOx). Whilst not as visible to the naked eye as PM emitted by diesel engines, gasoline engines produce PM of the size-order of $< 1\mu m$ such as 10-100nm. Interest in gasoline PM is growing as particles of this size can penetrate deep into the human lungs and can be detrimental to health.

The amount of these pollutants that can be emitted by vehicular internal combustion engines is prescribed by legislation in various countries and regions of the world, such as the USA and Europe, and these amounts are set to decrease as the legislation tightens step-wise over the next ten years or so. Similarly, International agreements between countries have led to moves toward vehicular internal combustion engines that use fuel more efficiently. The legislation acts as a stimulus to vehicle manufacturers and to their suppliers to devise new engines that are more fuel-efficient and that emit fewer pollutants and to exhaust systems that can clean up the exhaust gas before it passes to atmosphere.

One such exhaust system component primarily for treating diesel exhaust comprises an oxidation catalyst for oxidising NO in the exhaust gas to NO_2 and a downstream filter for trapping PM. A process for treating diesel PM that uses this arrangement is described in EP-B-0341382 or US-A-4,902,487, both of which are incorporated herein by reference. The process comprises passing an exhaust gas, such as a diesel exhaust gas, including PM and NO unfiltered over an oxidation catalyst to convert the NO to NO_2, collecting soot on the filter and combusting the collected soot by reaction with the NO_2. This technology is commercially available as Johnson Matthey's Continuously Regenerating Trap or CRT™. Further steps may be added, for example

downstream NOx removal by injection of reductant e.g. HC or NOx-specific reactant e.g. NH_3 or urea (see for example our WO-A-00/21647). An advantage of this process is that it is possible to combust diesel soot at temperatures of up to 400°C, whereas combustion of diesel soot in oxygen occurs at about 500°C. This is significant since diesel exhaust gas is generally cooler than exhaust gas from gasoline engines and soot would accumulate on the filter causing back-pressure problems in the system if the process relied on combustion of soot in oxygen.

One form of gasoline engine is a gasoline direct injection engine, which is designed to operate under stoichiometric and lean conditions. When running lean, relatively low levels of NOx are formed that cannot be reduced (removed) in the presence of the relatively high levels of oxygen in the exhaust gas. Reducing species, e.g. HC, can reduce NOx to N_2 during stoichiometric- or rich-running conditions, as comparatively less oxygen is present than during lean-running conditions.

In order to control NOx in lean-burn engines, there has been devised a NOx absorber/catalyst which can store NOx, e.g. as nitrate, when an engine is running lean. In a stoichiometric or rich environment, the nitrate is understood to be thermodynamically unstable, and the stored NOx is released and is reduced by the reducing species present in the exhaust gas. This NOx absorber/catalyst is commonly called a NOx-trap and is described in EP-A-0560991. By periodically controlling the engine to run stoichiometrically or rich, stored NOx is reduced and the NOx-trap regenerated.

A typical NOx-trap formulation includes a catalytic oxidation component, such as platinum, a NOx-storage component, such as barium, and a reduction catalyst e.g. rhodium. One mechanism commonly given for NOx-storage during lean engine operation for this formulation is:

(i) $NO + 1/2O_2 \rightarrow NO_2$; and

(ii) $BaO + NO_2 + 1/2O_2 \rightarrow Ba(NO_3)_2$.

In the first step, the nitric oxide reacts with oxygen on active oxidation sites on the platinum to form NO_2. The second step involves adsorption of the NO_2 by the storage material in the form of an inorganic nitrate.

When the engine runs under rich conditions or at elevated temperatures, the nitrate species become thermodynamically unstable and decompose, producing NO or NO_2 according to equation (iii) below. Under rich conditions, these nitrogen oxides are subsequently reduced by carbon monoxide, hydrogen and hydrocarbons to N_2, which can take place over the reduction catalyst.

(iii) $Ba(NO_3)_2 \rightarrow BaO + 2NO + 3/2O_2$ or $Ba(NO_3)_2 \rightarrow BaO + 2NO_2 + 1/2O_2$; and

(iv) $NO + CO \rightarrow 1/2N_2 + CO_2$ (and other reactions).

In the reactions of (i)-(iv) above the reactive barium species is given as the oxide. However, it is understood that in the presence of air most of the barium is in the form of the carbonate or possibly the hydroxide. The above reaction schemes can be adapted accordingly for species of barium other than the oxide.

Using sophisticated engine management techniques to provide for rich/lean cycling and common rail fuel injection, vehicle manufacturers are now adopting NOx-trap technology into diesel exhaust systems. One such system is described in EP-A-0758713. Means reducing the redox composition of the exhaust gas, as defined by lambda, for the purpose of regenerating a NOx-trap include injecting HC into the exhaust gas downstream of the engine, adjusting the ignition timing of at least one engine cylinder or adjusting the engine air-to-fuel ratio.

Another technique that can be used to control emissions is exhaust gas recirculation (EGR). In this, a portion of the exhaust gas is taken returned to the engine air intake so that the engine is fed a mixture of air and exhaust gas. Because the resulting mixture is lower in oxygen than in air, the temperature of the combustion event is reduced so that there is less NOx in the exhaust gas. This technique does cause an increase in PM, so there is a pay-off between NOx and PM, but by managing the rate of EGR to the load on the engine, it is possible to obtain an overall reduction in pollutant emissions.

In Japanese patent no. 2722987 and EP-A-1079084, Toyota describes an exhaust system including a component including a combination of certain of the catalyst features described above. Essentially it describes a catalysed particulate trap including a

NOx-trap. In particular, the component comprises a particulate trapping device comprising a NOx absorbent capable of absorbing NOx contained in exhaust gas when the air-fuel ratio of the exhaust gas is lean, and capable of releasing the NOx absorbed in the NOx component when the air-fuel ratio of the exhaust gas is substantially equal to the stoichiometric air-fuel ratio or rich.

The mechanism suggested for the combustion of soot trapped on the particulate trap is that during lean running, a high concentration of oxygen O_2 is deposited in the form of O_2^- or O^{2-} on the surface of platinum (Pt). NO contained in the flowing exhaust gas reacts with O_2^- or O^{2-} on the surface of the Pt to form NO_2 ($2NO + O_2 \rightarrow 2NO_2$). Then, part of the NO_2 thus formed is absorbed into the NOx absorbent while being oxidised on Pt, and diffused in the form of nitrate ion NO_3^- while combining with BaO.

If the air-fuel ratio is adjusted rich, the oxygen concentration in the exhaust gas is reduced, and consequently the amount of NO_2 formed on the surface of the Pt is reduced. If the amount of NO_2 produced is reduced, the reaction proceeds in the reverse direction ($NO_3^- \rightarrow NO_2$) and thus the nitrate ion NO_3^- is released in the form of NO_2 from the absorbent.

The suggestion is that "activated oxygen" species such as O_2^- and O^{2-} are responsible for combusting particulate during rich and lean running, but also that NO_2 could also be responsible for combustion of particulate, particularly during rich running.

We have investigated Toyota's combined particulate filter-NOx trap and have found, very surprisingly, that by introducing an oxidation catalyst active for oxidation of NO to NO_2 upstream of the filter/trap in a similar arrangement to that described in EP-B-0341832 or US-A-4,902,487 that filter regeneration is improved compared with filter regeneration employing the particulate filter-NOx trap alone. We have been able to show this by measuring the back-pressure in the system on a bench mounted engine. Increased back-pressure is an indication of increased particulate build up, i.e. that particulate deposition and particulate combustion are not in balance. It is also believed that the system represents an improvement over the system described in EP-A-758713 in that NOx released from the NOx absorbent can combust trapped particulate, but also oxidise HC to carbon dioxide (CO_2) and water (H_2O) and oxidise carbon monoxide CO

to CO_2. Accordingly, the system provides an improved management of pollutant species in the exhaust gas.

According to the invention, there is provided an exhaust system for an internal combustion engine, which system comprising a catalysed particulate filter comprising a NOx absorbent capable of absorbing NOx contained in an exhaust gas when the composition of the exhaust gas is lambda > 1, and capable of releasing the NOx absorbed in the NOx component when the exhaust gas composition is 1 ≥ lambda, characterised in that the exhaust system further comprises a catalyst capable of oxidising NO to NO_2 at least when the air-fuel ratio of the exhaust gas is lean.

We believe that an exhaust system including the particulate filter-NOx trap alone is less active for particulate combustion because the combustion of trapped particulate occurs only where it is in contact with the Pt or other washcoat components. Accordingly, particulate further from the surface of the filter-trap is combusted later than that which is nearer the surface. In the present invention particulate in contact with the Pt on the trap can be combusted at the same time as particulate not in contact with the Pt, because the particulate not in contact with the Pt is combusted in exhaust gas including increased levels of NO_2 downstream of the oxidation catalyst.

The invention is advantageous in that by reducing back-pressure in the system, fuel economy is improved and wear on the engine is reduced or eliminated.

Known catalysts for producing NO_2 from NO and O_2 may be used to generate the NO_2 oxidant for the purpose of combusting particulate. Such catalysts are extensively used in the catalytic conversion of automotive exhaust gases. This includes, for example, Pt, rhodium (Rh), ruthenium (Ru), palladium (Pd) or combinations thereof, platinum group metal oxides such as RhO_3 and the like. Conveniently, the catalyst is coated onto a monolith substrate e.g. a ceramic or metal honeycomb.

The filter may be in conventional form and structure. Typically this comprises a ceramic wall-flow filter of appropriate pore size, but one or more wire meshes of appropriate metal e.g. stainless steel or the like can also be used.

The NOx absorbent includes alumina, for example as a support, and at least one selected from, for example, alkali metals, such as potassium (K), sodium (Na), lithium (Li) and caesium (Cs), alkaline earth metals, such as barium (Ba) and calcium (Ca), and rare earth metals, such as lanthanum (La) and yttrium (Y), and a noble metal such as Pt carried on the support. A reduction catalyst such as rhodium can also be included.

According to a further aspect, the invention provides an internal combustion engine including an exhaust system according to the invention. The engine can be a diesel engine, such as a heavy duty diesel engine (as defined by the relevant European or US Federal or California State legislation) or a diesel engine for a light duty diesel engine, such as for a passenger vehicle or van. The engine can also be a gasoline engine, such as a lean-burn gasoline engine including a gasoline direct injection engine. However, the engine can be powered by alternative fuel means such as CNG, LPG or methanol, and engines powered by these alternative fuels are within the scope of the present invention.

In a further aspect, the invention comprises a vehicle including an internal combustion engine according to the invention. However, the exhaust system can be also be used in connection with stationary power plants.

According to a further aspect, the invention provides a method of treating an exhaust gas of an internal combustion engine, which method comprising oxidising NO in the exhaust gas to NO_2, trapping particulate on a catalysed filter also including a NOx absorbent, oxidising NO to NO_2 on the filter when the composition of the exhaust gas is lambda > 1, absorbing the NO_2 in the NOx absorbent when the composition of the exhaust gas is lambda > 1, releasing the absorbed NOx as NO_2 when the exhaust gas composition is 1 ≥ lambda and combusting particulate trapped on the filter in NO_2, optionally at exhaust gas temperatures of up to 400°C.

In order that the invention may be more fully understood, the following Example is provided by way of illustration only and with reference to the accompanying Figure which shows a graph depicting the effect of pre-NO oxidation catalyst and combined NOx and particulate trap.

EXAMPLE

A diesel particulate wall-flow filter (5.66 inches (14.38 cm) diameter by 6 inches (15.24 cm) long, 200 cells per square inch (31 cells cm^{-2})) was coated with a conventional NOx trap composition comprising supported platinum and barium prepared using known incipient wetness solution impregnation and conventional coating techniques. The coated filter was dried in an airflow and calcined at 500°C.

The resulting piece, now termed a NOx particulate trap (NPT), was mounted in a stainless steel can using standard procedures, and fitted to the exhaust gas system of a bench-mounted 1.9 litre common rail diesel engine. The engine was coupled to a dynamometer in the conventional manner, with both engine and dynamometer being controlled by computer. Exhaust emissions at pre- and post-NPT positions were measured at 10 second intervals. Gas pressures and temperatures at pre- and post-NPT positions were measured over the same time interval.

The engine was operated to give cycles of lean-running and rich-running conditions. The engine was run at 2300 rpm and the torque was adjusted to give a NPT gas inlet temperature of 350°C. After 60 seconds of lean-running the engine conditions were changed to rich conditions for 2 seconds by means of fuel post-injection, air intake throttling, and increased exhaust gas recirculation (EGR) rate. After two hours of cycling 60 seconds lean and 2 seconds rich the engine was kept at lean-running conditions and the torque was increased to give a NPT gas inlet temperature of 450°C. These lean-running conditions were maintained for 1 hour. The reaction between soot and NO_2 during this period was monitored by the reduction in back pressure of the system.

The above test conditions were repeated on a combined system comprising of diesel oxidation catalyst (DOC) followed by a NPT filter.

The DOC was prepared by coating a cordierite monolith (5.66 inches (14.38 cm) diameter by 3 inches (7.62 cm) long, 400 cells per square inch (62 cells cm^{-2})) with platinum supported on alumina using conventional coating techniques. The DOC was

mounted in a stainless steel can and fitted to the exhaust gas system of the diesel engine. The NPT filter was then fitted 1 inch (2.54cm) behind the DOC. Emissions and back pressure measurements were carried out over the lean-rich cycling and lean only conditions detailed above.

As can be seen from Figure 1, during the rich-lean cycling, the back-pressure in the system including the DOC upstream of the NPT is consistently lower than the back-pressure in the system without the DOC. Furthermore, it can be seen that following the switch to constant lean running, NO_2 increases downstream of the NPT in both systems. This is because the NOx absorbent is "full" or substantially all the NOx absorbent is in the nitrate form. With no rich regeneration events to reduce the nitrate and regenerate the NOx absorbent, the system including the DOC + NPT essentially becomes a CRT as described in EP-B-341832. NO_2 generated over the Pt of the NOx trap on the NPT appears to be responsible for the combustion of particulate on the NPT only system. In both cases, increased NO_2 is detected downstream of the NPT.

CLAIMS:

1. An exhaust system for an internal combustion engine, which system comprising a catalysed particulate filter comprising a NOx absorbent capable of absorbing NOx contained in an exhaust gas when the composition of the exhaust gas is lambda > 1, and capable of releasing the NOx absorbed in the NOx component when the exhaust gas composition is 1 ≥ lambda, characterised in that the exhaust system further comprises a catalyst capable of oxidising NO to NO_2 at least when the air-fuel ratio of the exhaust gas is lean.

2. An exhaust system according to claim 1, wherein the particulate trap is a ceramic wall flow filter.

3. An exhaust system according to claim 1 or 2, wherein the NOx absorbent comprises at least one of: an alkali metal, an alkaline earth metal and a rare earth metal.

4. An exhaust system according to claim 1, 2 or 3, wherein the filter further comprises an oxidation catalyst.

5. An exhaust system according to claim 4, wherein the oxidation catalyst is platinum and/or palladium.

6. An exhaust system according to any preceding claim, wherein the filter further comprises rhodium.

7. An exhaust system according to any preceding claim wherein the catalyst capable of oxidising NO to NO_2 at least when the air-fuel ratio of the exhaust gas is lean comprises a platinum group metal such as platinum and/or palladium.

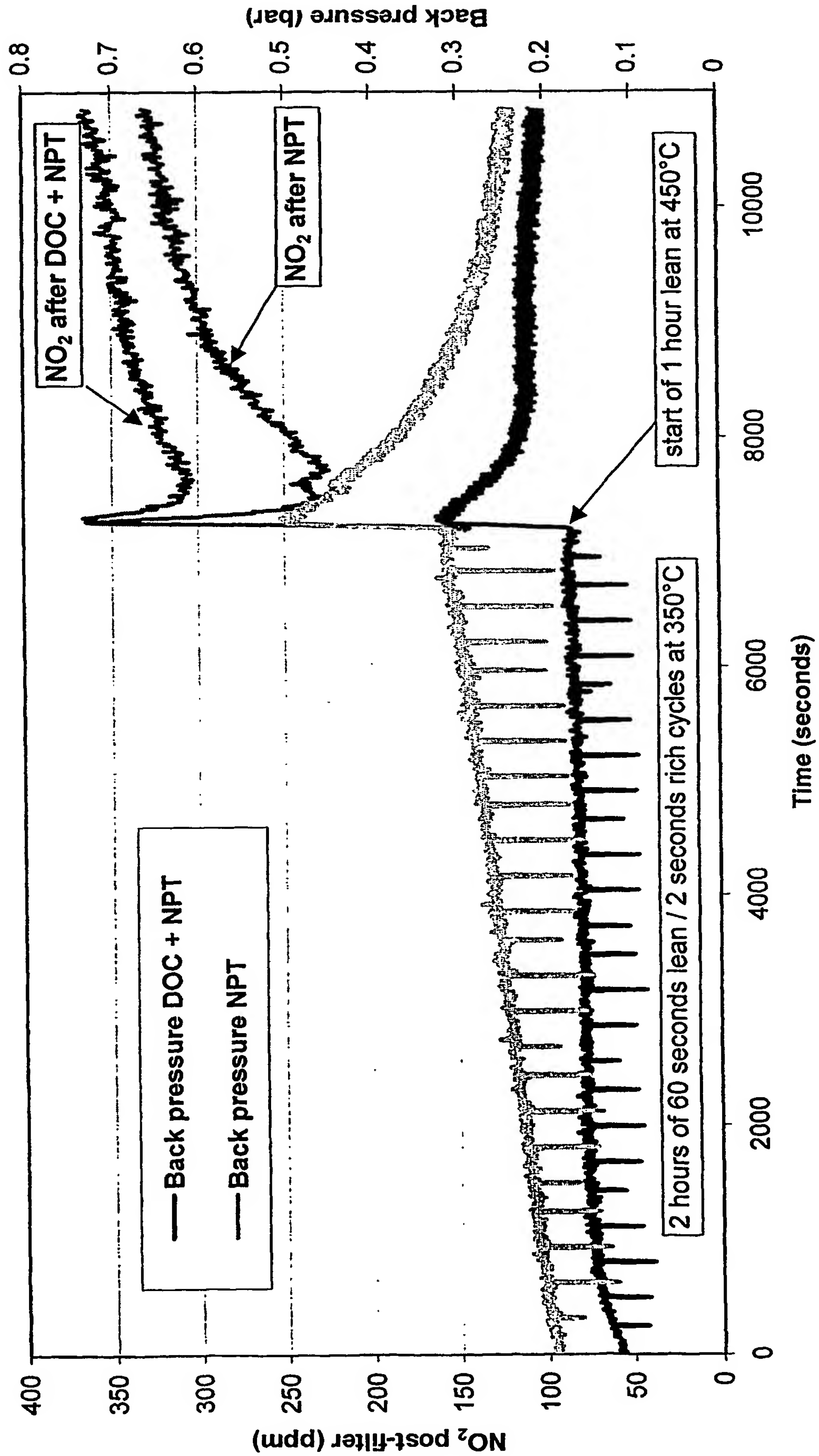
8. An internal combustion engine including an exhaust system according to any preceding claim.

9. An engine according to claim 8, wherein it is a diesel engine, such as a heavy duty diesel engine.

10. A vehicle including an internal combustion engine according to claim 8 or 9.

11. A method of treating an exhaust gas of an internal combustion engine, which method comprising oxidising NO in the exhaust gas to NO_2, trapping particulate on a catalysed filter also including a NOx absorbent, oxidising NO to NO_2 on the filter when the composition of the exhaust gas is lambda > 1, absorbing the NO_2 in the NOx absorbent when the composition of the exhaust gas is lambda > 1, releasing the absorbed NOx as NO_2 when the exhaust gas composition is $1 \geq$ lambda and combusting particulate trapped on the filter in NO_2, optionally at exhaust gas temperatures of up to 400°C.

Fig 1.


Back pressure (bar)
NO$_2$ after DOC + NPT
NO$_2$ after NPT
start of 1 hour lean at 450°C
2 hours of 60 seconds lean / 2 seconds rich cycles at 350°C
Back pressure DOC + NPT
Back pressure NPT
Time (seconds)
NO$_2$ post-filter (ppm)
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0
50
100
150
200
250
300
350
400
0
2000
4000
6000
8000
10000

INTERNATIONAL SEARCH REPORT

PCT/GB 02/04750

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 B01J23/38 B01J20/04 F01N3/08 B01D53/94

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 B01J F01N B01D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P,X	WO 02 18753 A (JOHNSON MATTHEY PLC ;SWALLOW DANIEL (GB); BRISLEY ROBERT JAMES (GB) 7 March 2002 (2002-03-07) abstract page 1, line 28 -page 2, line 21 page 3, line 2 - line 22 page 4, line 9 - line 16 page 5, line 13 - line 25 page 7, line 11 - line 13 page 9, line 26 -page 11, line 14; tables figures 3-10 ——— -/--	1,3-9

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
16 January 2003	24/01/2003

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL – 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Gosselin, D

INTERNATIONAL SEARCH REPORT

PCT/GB 02/04750

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication,where appropriate, of the relevant passages	Relevant to claim No.
X	WO 00 21647 A (JOHNSON MATTHEY PLC ;TWIGG MARTYN VINCENT (GB)) 20 April 2000 (2000-04-20) cited in the application page 2, line 1 -page 4, line 16 page 7, line 7 - line 8 claims 17,18	1-9
Y		2
X	DE 196 36 041 A (VOLKSWAGENWERK AG) 12 March 1998 (1998-03-12) column 1, line 60 - line 64 column 2, line 39 -column 3, line 9 column 3, line 65 -column 4, line 50 column 5, line 34 - line 68 column 6, line 54 -column 7, line 7 column 7, line 32 - line 34 claims; figure 1	1-9
Y		2
X	EP 1 033 161 A (DORNIER GMBH) 6 September 2000 (2000-09-06) paragraphs ‘0001!,‘0007!-‘0009!,‘0012!,‘0013!,‘0016!, ‘0017!,‘0019!,‘0021! claims 1,6,8,9 figure 1	1,5,7-9
Y		2
X	DE 199 41 439 A (RENAULT BOULOGNE BILLANCOURT) 16 March 2000 (2000-03-16) column 3, line 49 - line 57 column 4, line 25 - line 37 figures	1,8,9
Y		2
Y	EP 0 984 146 A (TOYOTA MOTOR CO LTD) 8 March 2000 (2000-03-08) paragraphs ‘0014!,‘0019!,‘0049!,‘0062!	2
X	EP 0 893 154 A (VOLKSWAGENWERK AG) 27 January 1999 (1999-01-27) column 1, line 1 - line 6 column 1, line 39 - line 48 column 3, line 6 - line 55 claims 10,11 figure 1	1,3-9

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INTERNATIONAL SEARCH REPORT

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 758 713 A (TOYOTA MOTOR CO LTD) 19 February 1997 (1997-02-19) cited in the application column 4, line 16 - line 24 column 4, line 50 -column 5, line 9 column 5, line 44 -column 6, line 10 column 6, line 49 - line 58 figure 1	1,3-9
A	EP 1 079 084 A (TOYOTA MOTOR CO LTD) 28 February 2001 (2001-02-28) cited in the application paragraph '0064! claim 1	
A	EP 1 008 379 A (INST FRANCAIS DU PETROL) 14 June 2000 (2000-06-14) abstract paragraphs '0010!,'0018!,'0021!,'0036!,'0063!	1,3,8,9

Form PCT/ISA/210 (continuation of second sheet) (July 1992)

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

Continuation of Box I.2

a. The present examination is solely carried out for the compositions supported by the description of the application, i.e. an exhaust line for internal combustion engine comprising as separated means an oxidation catalyst and a NOx absorber, the oxidation catalyst being upstream of the NOx absorber (i.a. page 4, line 21 to page 5, line 2 and examples). The search authority agrees with the objection put forward by the search authority as to a lack of support (Article 6 PCT) and disclosure (Article 5 PCT) for the extremely large number of possible catalysts, which have not been identified.

For the sake of completeness, it is submitted that even if embodiments, which have not been searched would be novel, it would not be possible to recognise an inventive step due to the absence of technical evidence that the claimed catalysts individually solve a technical problem or provide a technical effect. A generalisation of the results obtained with the embodiment disclosed in the application to other devices falling within the definition of claims 1 to 9 can therefore not be accepted.

b. Some of the features in the apparatus claim 1 relate to a method of using the apparatus rather than clearly defining the apparatus in terms of its technical features. The intended limitations are therefore not clear from this claim, contrary to the requirements of Article 6 PCT.

c. The order of the claims should be grouped so as to comply with the requirements of Rule 6.4c PCT. Claims 2,4 and 6 refer to the NOx absorber and claims 3, 5 and 7 to the oxidation catalyst.

The applicant's attention is drawn to the fact that claims, or parts of claims, relating to inventions in respect of which no international search report has been established need not be the subject of an international preliminary examination (Rule 66.1(e) PCT). The applicant is advised that the EPO policy when acting as an International Preliminary Examining Authority is normally not to carry out a preliminary examination on matter which has not been searched. This is the case irrespective of whether or not the claims are amended following receipt of the search report or during any Chapter II procedure.

INTERNATIONAL SEARCH REPORT

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Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☒ Claims Nos.:
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:
see FURTHER INFORMATION sheet PCT/ISA/210

3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.

2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.

3. ☐ As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:

4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

☐ The additional search fees were accompanied by the applicant's protest.

☐ No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

PCT/GB 02/04750

Patent document cited in search report		Publication date	Patent family member(s)			Publication date
WO 0218753	A	07-03-2002	AU	8235301	A	13-03-2002
			WO	0218753	A1	07-03-2002
WO 0021647	A	20-04-2000	AU	6111799	A	01-05-2000
			EP	1128895	A1	05-09-2001
			WO	0021647	A1	20-04-2000
DE 19636041	A	12-03-1998	DE	19636041	A1	12-03-1998
			WO	9810177	A1	12-03-1998
			EP	0925431	A1	30-06-1999
EP 1033161	A	06-09-2000	DE	19908023	A1	31-08-2000
			EP	1033161	A1	06-09-2000
DE 19941439	A	16-03-2000	FR	2783280	A1	17-03-2000
			DE	19941439	A1	16-03-2000
EP 0984146	A	08-03-2000	JP	2000073817	A	07-03-2000
			JP	2000087732	A	28-03-2000
			DE	19937952	A1	02-03-2000
			EP	0984146	A2	08-03-2000
			US	6233925	B1	22-05-2001
EP 0893154	A	27-01-1999	DE	19731623	A1	28-01-1999
			EP	0893154	A2	27-01-1999
EP 0758713	A	19-02-1997	JP	9053442	A	25-02-1997
			DE	69612645	D1	07-06-2001
			DE	69612645	T2	18-04-2002
			EP	0758713	A1	19-02-1997
			US	5746989	A	05-05-1998
EP 1079084	A	28-02-2001	JP	3304929	B2	22-07-2002
			JP	2001065330	A	13-03-2001
			EP	1079084	A2	28-02-2001
EP 1008379	A	14-06-2000	FR	2787037	A1	16-06-2000
			EP	1008379	A1	14-06-2000
			JP	2000170523	A	20-06-2000